MONITORING A SAMPLE CONTAINING A NEUTRON SOURCE

This invention is concerned with improvements in and relating to monitoring, in particular, but not exclusively, monitoring of nuclear materials.

Monitoring of nuclear waste, and plutonium waste in particular, is important both for accountancy purposes and in ensuring that criticality safety standards are maintained. It is important to maintain an inventory of the plutonium in a plant and during disposal, for instance, to know the exact amount of plutonium in a given volume of waste.

Determination of the mass of plutonium in a waste sample is presently conducted using neutron coincidence counting (NCC). On occasions it is necessary to supplement these investigations with high resolution gamma spectrometry measurement to check the validity of assumptions on the isotopic make up of the sample. For waste samples such as drums, boxes and other handleable packages a detection chamber is normally constructed into which the samples to be measured are introduced.

The detection chamber principally consists of a series of neutron detecting tubes placed in a polyethylene moderating material. The signals produced by the detectors are analysed electronically to determine those signals attributable to spontaneous fissions of the plutonium rather than background radiation and other sources. The spontaneous fissions of the plutonium produce 2, 3, 4 and higher numbers of neutrons for each event. The spontaneous fission rate is known to be proportional to the mass of certain isotopes and so to the neutron source material as a whole. Signals detected within a certain time window are taken as being indicative of a pair, rather than a single neutron, which can neither be

attributed to the background or the plutonium with any
certainty.

As the component of the signal relating to the pairs is
indicative of the plutonium mass of certain isotopes,
principally ^{240}Pu then by making certain assumptions about the
isotopic makeup of the sample the total mass can be
suggested.

As well as requiring such assumptions as to the material
makeup which are inappropriate given the likely variation
over time of the isotopes in the waste, NCC also suffers from
significant errors or inaccuracies due to the variable
shielding effects of the different waste materials in which
the plutonium of interest may be distributed as well as due
to the different materials associated with the containers for
the waste. For instance water, which will be present to
varying degrees in the waste, and polyethylene or PVC, which
may be associated with the container, are each strong neutron
moderating materials. Variation in any of these can
drastically effect the detection efficiency and hence the
result. Problems with the effects of changes in the
detection efficiency due to the materials actual position
within the sample and as a consequence within the chamber are
also encountered.

As an improvement it has been proposed to undertake
multiplicity counting (MC) in which the events producing
single and triplet events are considered alongside the
doublets. However, the count rates arising relate to four
unknown parameters; the spontaneous fission rate (reflecting
the isotopic make up of the material); the self-induced
fission are (known as multiplication); the detection
efficiency; and the α,n reaction rate. Several other nuclear
constants are also involved. The presence of three

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experimentally determinable factors, the single, double and
triple count rates, still leaves the system unsolvable as
there are four unknowns. Detection of the quadlet counting
rate is impracticable due to the very limited number of
detectable occurrences for such events. In even the best
prior art systems, therefore, it is necessary to set one of
the unknowns at a predetermined value in reaching a solution.

Attempts to set any one of the self-induced fission rate;
detection efficiency; or α,n reaction rate are not accurate
in waste samples.

The self-induced fission rate varies due to variations in
materials and locations with the result that even a small
error has a substantial effect on the end result due to the
factors role in the equations. The detection efficiency
differs from that obtained in calibration due to variations
in the location of the plutonium in the waste sample and due
to variations within the accompanying materials. The α,n
rate changes with the isotopic, chemical and materials in the
waste matrix which will vary between samples. In each case
errors in the end result arise resulting in inefficient and
costly waste disposal schemes or an inaccurate inventory.

According to a first aspect of the invention we provide a
method of monitoring a sample containing a neutron source in
which:-

        i) signals from a plurality of neutron detectors are
analysed and the count rates for single, double and triple
incidence of neutrons on the detectors are determined;

        ii) the single, double and triple count rates are
equated to a mathematical function related to the spontaneous
fission rate, self-induced fission rate, detection efficiency
and α,n rate;

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iii) a probability distribution is assigned to each of the self-induced fission rate, detection efficiency and α,n reaction rate and each of the counting rates to provide a probability distribution factor for any given value;

iv) and the value of the product of all the probability distribution factors is increased to give an optimised solution and so provide a value for the spontaneous fission rate which is linked to the mass of the neutron source.

In this way an accurate value for the mass of a neutron source within a sample of material is provided without having to make hard assumptions about any of the variables within the system.

Quadruplet or high counts could be substituted for one of the single, pair of triple counts, but the number of events / unit time would be reduced accordingly.

The sample may be in the form of a piece of equipment, such as a glove box, ventilation unit or the like; a component of a piece of equipment, such as a filter; or a container, such as a drum, box or other package. The container may be sealed. The sample may be of waste material, such as material destined for disposal, or it may be material with a future active life for which an inventory is required.

The sample may include other materials besides the neutron source. For instance other materials such as non-neutron source elements, metals or compounds, water, plastics, glass and other sealing materials may be present.

The neutron source may comprise one or more elements or compounds, one or more isotopes of an element or mixtures of both. The neutron source may be naturally occurring and/or

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arise from fission reaction products. Plutonium and ^{240}Pu in particular are neutron sources which may require such monitoring.

The neutron detectors may be of the ^{3}He type. The detectors maybe provided in polyethylene or other hydrogen providing material. In this way the neutron source to be monitored can be controlled and the neutrons moderated to detectable energy levels. Alternative or additional neutron absorbing materials, such as boron or cadmium, may be provided to shield the detectors, for instance by positioning around the detectors, against background events.

The detectors may be provided around all sides and most preferably above and/or below the sample during monitoring. Between 20 and 125 detectors may be provided. Preferably between 30 and 50 detectors are provided around the sides of the sample. Preferably between 8 and 16 detectors are provided above and below the sample.

Preferably each detector is provided with an amplifier (generally to increase the pulse to a signal level suitable for further processing) and a discriminator (generally to reject or eliminate noise events.

The detector signals are preferably summed and fed to sequence analysing means. Preferably each pulse causes a time period to be considered, with other pulses being received in that period being associated with the initial pulse. In this way sequences of single, double, triple and greater numbers of neutron detections are obtained. Neutrons detected within the time period are accepted as originating from the same spontaneous fission within the sample as the initial pulse. Preferably the detections are subjected to a correction factor. The correction factor may account for

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accidental coincidences, for instance detections from
different source simultaneous emissions which give the
impression of a pair or greater.

Preferably the time period lasts between 10 and 510 micro
seconds and most preferably between 50 and 100 micro seconds.

Preferably the start of the time period occurs between 5 and
10 micro seconds after the initial pulse.

Preferably the singlet count rate is related to the
spontaneous fission rate, the self-multiplication factor,

where m = $\dfrac{1-p}{(1-p)\upsilon_I}$

and p = probability first neutron causes induced fission; the
detection efficiency and the α,n reaction rate by the
function

$$R_1 = \varepsilon . F_s . M . v_{s1} . (1+\alpha)$$

Preferably the doublet counting rate is related to the
spontaneous fission rate, the self-multiplication factor,
where

$$m = \frac{1-p}{(1-p)\upsilon_I}$$

and p = probability first neutron causes induced fission; the
detection efficiency and the α,n reaction rate by the
function

$$R_2 = \varepsilon^2 \cdot F_s \cdot M^2 \cdot v_{s2} \cdot \left(1 + (M-1)(1+\alpha)\frac{v_{s1}v_{I2}}{v_{s2}(v_{I1}-1)} \right)$$

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Preferably the triplet counting rate is related to the
spontaneous fission rate, the self-multiplication factor,
where

$$m = \frac{1-p}{(1-p)u_I}$$

and p = probability first neutron causes induced fission; the
detection efficiency and the α,n reaction rate by the
function

$$R_3 = \varepsilon^3 \cdot F_s \cdot M^3 \cdot v_{s3} \cdot \left(1 + 2(M-1)\frac{v_{s2}v_{I3}}{v_{s3}(v_{I1}-1)} + (M-1)(1+\alpha)\frac{v_{s1}v_{I3}}{v_{s3}(v_{I1}-1)}\left(1 + 2(M-1)\frac{v_{I2}^2}{v_{I3}(v_{s1}-1)}\right)\right)$$

Preferably the probability distribution assigned to
individual variables or counting rates is a normal
distribution or a flat distribution or a triangular
distribution. Normal distributions are preferably used for
one or more, and most preferably all, the counting rates.
Triangular distributions are preferably used for one or more,
and most preferably all, the individual variables, such as
detector efficiency, fission rate, multiplication
distribution and alpha distribution. A flat distribution is
preferably used for the fission rate.

The probability distributions may be symmetrical. The
probability distributions may be skewed.

The distribution may be constrained within certain applied
constraints / boundaries such that the probability
distribution factor is zero beyond the constraints,
particularly for triangular distributions.

The distribution may be modified such that the probability
distribution factor rapidly tends to zero beyond certain
values, particularly for normal distributions.

The constraints beyond which the distribution is zero or
values beyond which a rapid transition to zero occurs, may be
selected from one or more of the following :-

for the detection efficiency $0 \leq E \leq 1$;

for the self-induced fission rate $M \geq 1$;

for the α,n reaction $\alpha \geq 0$.

The ranges of the distributions of one or more of the single
count rate; the double count rate; and the triple count rate
are preferably based on the measured count rates and their
standard deviation.

The lower and / or upper values of the distributions, may be
provided according to one or more or all of the following
ranges :-

a) for the self-induced fission rate: according to the
anticipated plutonium content; between 1.0 and 1.3,
preferably 1.0 and 1.2; particularly for a high plutonium
content (ie, >100g Pu) between 1.0 and 1.2; particularly for
a low plutonium content (ie. <10g Pu) between 1.00 and 1.01;
and for intermediate plutonium contents, intermediate ranges;

b) for the detection efficiency: according to the
anticipated moderator content; between 0 and 0.3; between 0
and 0.15 for anticipated maximum efficiency levels; between
0.05 and 0.15 for moderator containing materials;

c) for the alpha,n reaction rate: according to the form
or forms of plutonium anticipated; between 0 to 10, where
plutonium fluorides are anticipated as present; between 0 to
2, or 0 to 1 where plutonium metal is anticipated; ;greater
than 0.0 where no plutonium metal is anticipated, for
instance between 0.4 and 1.0 or 0.5 and 1.0.

One or more of the constraints may be set according to
information gathered from a preceding isotopic consideration

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or analysis of the sample. Gamma spectrometry may be
employed. Information about the presence of plutonium metal
and / or plutonium oxides and / or plutonium fluorides and /
or moderators and / or other species or components may be
obtained and applied.

The permissible values for the spontaneous fission rate may
be constrained. Preferably the spontaneous fission rate is
constrained to $F \geq 0$.

The distributions may be provided according to the general
form :

$$pdf(\epsilon) = \frac{1}{\sqrt{2\pi}} \exp\left[-\tfrac{1}{2}.\left(\frac{\epsilon - \mu_\epsilon}{\sigma_\epsilon}\right)^2\right]$$

The initial set of values assigned to the variables may be
selected by the operator, predetermined or a function of
prior samples which have been analysed.

The increasing, and preferably maximisation, of the product
of the probability distribution factors (pdf's) is preferably
performed as an iterative process. The values of one, two,
three and most preferably all four of the variables are
varied in each iteration. The variation or correction
applied may be of a fixed value between iterations but a
variable correction is preferred.

One or more of the probability distribution factors and /or
the corresponding level of the variable from an optimised or
maximised solution may be used to redefine the range of the
applicable distribution used. One or more redefined
distributions may be used in a subsequent optimisation or
maximisation according to the method. Tighter errors apply
as a result as tighter constraints would be applied. The

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redefining of the distribution(s) and their use in
optimisation may be performed repeatedly.

The correction may be a correction vector. Most preferably
the correction vector is derived from d(pfd)/d(x) where x is
the variable, solved for zero.

In one method , because there are four variables, the part
function may be partially differentiated to yield a set of
four equations:-

eg. $\mu_E = 0.12$ $\sigma_E = 0.03$

       $\mu_m = 1.02$ $\sigma_m = 0.02$

       $\mu_\alpha = 0.4$ $\sigma_\alpha = 0.1$

The product pdf may be partially differentiated to provide
four simultaneous equations. The equations may be

$$\frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial \varepsilon}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial F_s}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial M}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial \alpha}$$

Alternatively or additionally the effects of small variations
can be determined based on linearised equations in four
dimensions. The use of equations

$$\frac{\delta\left(\frac{\delta pdf}{\delta \varepsilon}\right)}{\delta \varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta \varepsilon}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta \varepsilon}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta \varepsilon}\right)}{\delta \alpha}\Delta\alpha = -\frac{\delta pdf}{\delta \varepsilon}$$

$$\frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta \varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta \alpha}\Delta\alpha = -\frac{\delta pdf}{\delta F_s}$$

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$$\frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta M}$$

$$\frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta\alpha}$$

where

$$\frac{\delta\left(\frac{\delta pdf}{\partial\varepsilon}\right)}{\partial F_s} \approx \frac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta F_s} = \frac{\left(\frac{\delta pdf(\varepsilon,F_s,M,\alpha)}{\delta\varepsilon} - \frac{\delta pdf(\varepsilon,F_s+\Delta F_s,M,\alpha)}{\delta\varepsilon}\right)}{\delta F_s}$$

is the 2nd order derivative of

$$\left(\frac{\partial pdf}{\partial\varepsilon}\right) \approx \left(\frac{\delta pdf}{\delta\varepsilon}\right) = \frac{pdf(\varepsilon+\delta\varepsilon,F_s,M,\alpha,\mu_{R_1},\mu_{R_2},\mu_{R_3}) - pdf(\varepsilon,F_s,M,\alpha,\mu_{R_1},\mu_{R_2},\mu_{R_3})}{\delta\varepsilon}$$

etc

may be employed. These functions may be solved to give a correction vector, for instance

$$\begin{bmatrix} \Delta_e \\ \Delta_F \\ \Delta_m \\ \Delta_\alpha \end{bmatrix}$$

The method may handle the solution in matrix form.

Preferably the process is repeated until the correction vector is below a given threshold. The threshold may be predetermined.

The initial pdf values for the allocated initial values are preferably evaluated as positive and negative correction vectors. Preferably each subsequent iterations pdf values

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are similarly evaluated. In this way the effects of poor
starting criteria are alleviated.

For a pdf the correction vector may be multiplied by a
constant factor and divided by a constant factor and the
corresponding new pdf compared with the preceding pdf, if the
pdf value is greater than the preceding pdf the new
correction vector is applied, if the pdf value is less than
the preceding value the new correction factor is divided by
the constant once more and the new pdf for this further
correction factor compared with the new preceding pdf, with
the stages further being repeated if necessary. Preferably
the multiplication factor is 32 and/or the divisive factor is
0.5. Most preferably these steps are performed for both
positive and negative correction vectors.

If one or more of the initially allocated values corresponds
to a pdf of zero a new value may be allocated. Alternatively
the count rate standard deviations maybe multiplied by a
constant factor, greater than 1. In this way a pdf value can
be provided for which derivatives can be calculated.
Preferably once the correction vector is below a certain
threshold the standard deviation can be divided by the
initial constant or a lower figure and the solution maximised
once more. This process is preferably repeated until the
inflationary factor is 1.

Preferably the method includes the provision of associated
error estimates. Total uncertainty within the system may be
determined.

The associated error estimates may be determined by equations

$$\left(\sigma_{\varepsilon}\right)^{2} = \left(\left(\frac{\partial \varepsilon}{\partial R_{1}}\right)\sigma_{R_{1}}\right)^{2} + \left(\left(\frac{\partial \varepsilon}{\partial R_{2}}\right)\sigma_{R_{2}}\right)^{2} + \left(\left(\frac{\partial \varepsilon}{\partial R_{3}}\right)\sigma_{R_{3}}\right)^{2}$$

$$\left(\sigma_{\hat{F}_{i}}\right)^{2} = \left(\left(\frac{\partial \hat{F}_{i}}{\partial R_{1}}\right)\sigma_{R_{1}}\right)^{2} + \left(\left(\frac{\partial \hat{F}_{i}}{\partial R_{2}}\right)\sigma_{R_{2}}\right)^{2} + \left(\left(\frac{\partial \hat{F}_{i}}{\partial R_{3}}\right)\sigma_{R_{3}}\right)^{2}$$

$$\left(\sigma_{\hat{M}}\right)^2 = \left(\left(\frac{\partial\hat{M}}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{M}}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{M}}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{\alpha}}\right)^2 = \left(\left(\frac{\partial\hat{\alpha}}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{\alpha}}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{\alpha}}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

Alternatively the associated error estimates may be obtained
from the equations

$$\left(\sigma_{\hat{\varepsilon}}\right)^2 = \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{F}_s}\right)^2 = \left(\left(\frac{\delta\hat{F}_s}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{F}_s}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{F}_s}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{M}}\right)^2 = \left(\left(\frac{\delta\hat{M}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{M}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{M}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{\alpha}}\right)^2 = \left(\left(\frac{\delta\hat{\alpha}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{\alpha}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{\alpha}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

where

$$\left(\frac{\partial\hat{\varepsilon}}{\partial R_1}\right) \approx \left(\frac{\delta\hat{\varepsilon}}{\delta R_1}\right) = \frac{\hat{\varepsilon}\left(\mu_{R_1}+\delta\mu_{R_1},\mu_{R_2},\mu_{R_3}\right) - \hat{\varepsilon}\left(\mu_{R_1},\mu_{R_2},\mu_{R_3}\right)}{\delta\mu_{R_1}}$$

etc.

where $\delta\mu_{R_1}$ = a small change in μ_{R_1} etc.
$\varepsilon(\mu_{R1}, \mu_{R2}, \mu_{R3})$ = final estimate from solution to $(\mu_{R1}, \mu_{R2}, \mu_{R3})$ count rates set.

According to a second aspect of the invention we provide:-
        i) apparatus for monitoring a sample containing a
neutron source comprising one or more neutron detectors;
        ii) processing means to receive signals from the
detectors; and wherein the signals are processed to provide a
count rate value indicative of the number of single, double
and treble incidences of neutrons on the detectors;

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wherein the counting rates are mathematically related to the spontaneous fission rate, self-induced fission rate, detection efficiency and α,n reaction rate;

wherein a probability distribution is assigned by the processing means to each of the self induced fission rate, detection efficiency and α,n reaction rate and the three counting rates; and

wherein the product of the probability distribution factors for values of the variables is increased by the processing means, the solution providing a value proportional to the mass of the neutron source present.

The sample may be in the form of a piece of equipment, such as a glove box, ventilation unit or the like; a component of a piece of equipment, such as a filter; or a container, such as a drum, box or other package. The container may be sealed. The sample may be of waste material, such as material destined for disposal, or it may be material with a future active life for which an inventory is required.

The sample may include other materials besides the neutron source. For instance other materials such as non-neutron source elements, metals or compounds, water, plastics, glass and other sealing materials may be present.

The neutron source may comprise one or more elements or compounds, one or more isotopes of an element or mixtures of both. The neutron source may be naturally occurring and/or arise from fission reaction products. Plutonium and ^{240}Pu in particular are neutron sources which may require such monitoring.

The neutron detectors may be of the ^{3}He type. The detectors maybe provided in polyethylene or other hydrogen providing material. In this way the neutron source to be monitored can

be controlled and the neutrons moderated to detectable energy
levels. Alternative or additional neutron shielding
materials such as boron or cadmium may be provided to shield
the detectors, for instance by positioning around the
detectors.

The detectors may be provided around all sides and most
preferably above and/or below the sample during monitoring.
Between 20 and 125 detectors may be provided. Preferably
between 30 and 50 detectors are provided around the sides of
the sample. Preferably between 6 and 16 detectors are
provided above and below the sample.

The detector signals are preferably summed and fed to
sequence analysing means. Preferably each pulse causes a
time period to be considered, with other pulses being
received in that period being associated with the initial
pulse. In this way sequences of single, double, triple and
greater numbers of neutron detections are obtained. Neutrons
detected within the time period are accepted as originating
from the same spontaneous fission within the sample as the
initial pulse.

Other features and options available for the apparatus and
its operating procedure are provide elsewhere within the
application.

According to a third aspect of the invention we provide a
sample monitored using the method of the first aspect of the
invention and/or using the apparatus of the second aspect of
the invention.

Various embodiments of the invention will now be described by
way of example only and with reference to the accompanying
drawings in which :-

Figure 1 is an illustration of a detection chamber layout;

Figure 2 illustrates a distribution assigned to the detection efficiency;

Figure 3a illustrates the probability distribution for a variable under consideration;

Figure 3b shows the partial derivative function and a correction vector for the probability distribution of Figure 4a;

Figure 4a illustrates the probability distribution for an alternative determination;

Figure 4b shows the partial derivative function for the probability distribution of Figure 5a illustrating the problem with poorly selected initial values;

Figure 5 provides a comparison of NCC and multiplicity counting measurements of a ^{252}Cf source in various matrix filled drums;

Figure 6 illustrates a comparison of NCC and multiplicity counting measurements of a Pu source in various matrix filled drums;

Figure 7 provides a comparison of NCC and multiplicity counting limits of detection for a drum containing 50kg of PVC; and

Figure 8 provides a comparison of NCC and multiplicity counting limits of detection for a drum containing 270kg of steel hulls.

The detection chamber illustrated in Figure 1 comprises a ten
sided chamber 2 with an openable wall portions 4 to provide
access for a drum 6 or the like to be monitored. Four ^{3}He
detectors are provided in each side with twelve similar
detectors in both the ceiling and floor 10 of the chamber.

Drums, 200 litre drums can be accommodated, may be lifted
into the chamber, positioned there by forklift truck or
conveyed on a bed of rollers. Suspending the drum is
preferred to avoid having to strengthen the base of the
chamber. Any such bearing surface could interfere with the
detection efficiency of the base detectors. Once positioned
the chamber is shut.

The neutron detectors are of conventional type in tube
configuration 1m long, 2.5cm in diameter and operated at 4
atmosphere pressure and 960V. The outside of the chamber 2
is provided with an 24cm thick layer of polyethylene to act
as a neutron shield to any background radiation. The
detectors are arranged vertically in bores in an inner layer
of 8 cm thick polyethylene. A 1mm layer of cadmium is
provided on the inside and outside of the layer to prevent
neutrons returning to the chamber and to prevent thermalise
neutrons reaching the detectors.

Each detector is provided with a preamplifier, amplifier and
discriminator to minimise dead time effects on the signals.
The signals are all summed and then fed to a frequency
analyser.

The frequency analyser works on each pulse in the following
manner. A received pulse opens after a preset period an
observation interval with the number of pulses falling within
this period being counted. A frequency distribution for the
number of pulses detected within the observation window is

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generated as a result. Singlets, doublets, triplets,
quadruplets and higher sets can be expected with decreasing
occurrences.

The singlet, doublet and triplet counting rates are
determined according to the following procedure.

The total neutron count rate observed during the measurement,
N_T, is obtained simply by counting the number of signals.
The first and second factorial moments of the frequency
distribution, $M_{m(1)}$ and $N_{m(2)}$, are also calculated from the
frequency table, using the following equations:-

$$N_{m(1)} = \sum_{x=1}^{x=N_{max}} x \cdot N_x \tag{1}$$

$$N_{m(2)} = \sum_{x=2}^{x=N_{max}} \binom{x}{2} \cdot N_x = \sum_{x=2}^{x=N_{max}} \frac{x(x-1)}{2} \cdot N_x \tag{2}$$

where: N_x is the normalised frequency to have x
        signals in the observation interval.
     N_{max} is the maximum number of signals observed in any
        observation interval.

The total count rate and the first and second factorial
moments are then background corrected and used in the
following equations to calculate the singlet count rate, and
the rate of correlated doublets and triplets, R_1, R_2 and R_3
respectively.

$$R_1 = N_T \tag{3}$$

$$R_2 = \frac{N_T \cdot N_{m(1)} - R_1^2 \cdot \tau}{f} \tag{4}$$

$$R_3 = \frac{N_T \cdot N_{m(2)} - R_2 \cdot R_1 \cdot \tau \cdot \{f + W_2\} - \frac{1}{2} \cdot R_1^3 \cdot \tau^2}{f^2} \tag{5}$$

where: τ = length of the observation interval

  $1/\lambda$ = neutron die away time

  T   = pre-delay

$$f = e^{-\lambda T}\left(1 - e^{-\lambda \tau}\right) \qquad (6)$$

$$W_2 = 1 - \frac{1}{\lambda\tau}\left(1 - e^{-\lambda\tau}\right) \qquad (7)$$

The counting rates obtained can then be applied to the following equations which relate the counting rates to the spontaneous fission rate, the self-induced fission rate, the detection efficiency and the alpha, n reaction rate.

$$R_1 = \varepsilon \cdot F_s \cdot M \cdot v_{s1} \cdot (1 + \alpha)$$

$$R_2 = \varepsilon^2 \cdot F_s \cdot M^2 \cdot v_{s2} \cdot \left(1 + (M-1)(1+\alpha)\frac{v_{s1}v_{I2}}{v_{s2}(v_{I1}-1)}\right)$$

$$R_3 = \varepsilon^3 \cdot F_s \cdot M^3 \cdot v_{s3} \cdot \left(1 + 2(M-1)\frac{v_{s2}v_{I2}}{v_{s3}(v_{I1}-1)} + (M-1)(1+\alpha)\frac{v_{s1}v_{I3}}{v_{s3}(v_{I1}-1)}\left(1 + 2(M-1)\frac{v_{I2}^2}{v_{I3}(v_{s1}-1)}\right)\right)$$

where ϵ = detector efficiency F_s = the spontaneous fission rate of the sample

  M   = the leakage multiplication

  α = (α, n) to spontaneous fission ratio

  n_{sn} = the nth spontaneous fission factorial moment

  n_{In} = the nth induced fission factorial moment

  v_{sn} = the nth spontaneous fission factorial moment
(for plutonium)

  v_{In} = the nth induced fission factorial moment (for plutonium)

Rather than fixing one of the unknown values to obtain a solution to the equations the present invention seeks a best fit solution to the information obtained based on certain

known absolute constraints and certain probability distributions assigned to each of the unknowns. Information about the mean values and standard deviations for the counting rate are determined experimentally. Information on the distributions for the other parameters can be obtained by calibration experiments aswell as being based on experience and past tests.

The distributions are provided in the following manner :-

Detection Efficiency Distribution

$$\varepsilon \sim N(\mu_\varepsilon, \sigma_\varepsilon)$$
$$0 \leq \varepsilon \leq 1$$

where μ_ε = mean value of the efficiency distribution
σ_ε = standard deviation of the efficiency distribution

Fission Rate Distribution

Is constrained to be $F_s \geq 0$ but is otherwise left free floating as the variable to be obtained.

Multiplication Distribution

$$M \sim N(\mu_M, \sigma_M)$$
$$M \geq 1$$

where μ_M = mean value of the multiplication distribution
σ_M = standard deviation of the multiplication distribution

Alpha Distribution

$$\alpha \sim N(\mu_\alpha, \sigma_\alpha)$$
$$\alpha \geq 0$$

where μ_α = mean value of the alpha distribution

σ_{α} = standard deviation of the alpha distribution

Singles Count Rate Distribution

$R_1 \sim N(\mu_{R1}, \sigma_{R1})$

where μ_{R1} = mean value of the singles distribution obtained by measurement

σ_{R1} = standard deviation of the singles distribution

Doubles Count Rate Distribution

$R_2 \sim N(\mu_{R2}, \sigma_{R2})$

where μ_{R2} = mean value of the doubles distribution obtained by measurement

σ_{R2} = standard deviation of the doubles distribution

Triples Count Rate Distribution

$R_3 \sim N(\mu_{R3}, \sigma_{R3})$

where μ_{R3} = mean value of the triples distribution obtained by measurement

σ_{R3} = standard deviation of the triples distribution

The probability distribution function (pdf) is assigned a normal distribution with the pdf of any trial value being calculated by

$$z = \left(\frac{y_i - \mu_y}{\sigma_y} \right)$$

$$pdf(y_i) = \frac{1}{\sqrt{2\pi}} \cdot \exp\left(-0.5 z^2\right)$$

Figure 2 illustrates such a normal distribution for the detection efficiency. Strictly speaking the pdf 20 should change abruptly to zero when either of the constraints for the efficiency is reached, i.e. greater than or equal to zero

and less than or equal to one, however, if such strict limits
are applied an adverse effect on the solution process
results. As a consequence a constrained pdf 22 is provided
but in a form which crosses the constraints to an extent.

The overall pdf for the variables can be determined by first
calculating the count rates and then determining the product
of all the individual pdf's. The solution is the set of
values for the unknown parameters which give the maximum pdf
product value, calculated according to

$$\text{pdf}(\varepsilon, F_s, M, \alpha, \mu 1_{R1}, \mu_{R2}, \mu_{R3}) = \text{pdf}(\varepsilon) \times \text{pdf}(F) \times \text{pdf}(M) \times \text{pdf}(\alpha)$$
$$\times \text{pdf}(\mu_{R1}) \times \text{pdf}(\mu_{R2}) \times \text{pdf}(\mu_{R3})$$

The method for determining the maximum solution to such a
function is to differentiate the function and solve for
d(function)/d(parameter)=zero. As the pdf product has four
input parameters it needs to be partially differentiated to
yield four simultaneous equations which can then be solved.
Linearisation method or Taylor series method is used to
determine corrections ($\Delta\varepsilon$; ΔF_{S1}; ΔM; $\Delta\alpha$) to be made to the
trial value set selected in order to reduce the observed
partial derivatives to zero.

Figure 3a shows a probability distribution 40 for a variable
with the partial derivative of this distribution being
provided in Figure 3b. This illustrates in a single
dimension the correction applied to an initial estimate 42 to
give the next estimate 44. This in turn produces a further
correction factor and a subsequent further estimate 46 and so
on.

In four dimensions the linearised equations are

$$\frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial \varepsilon}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial s}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial F_s}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial F_s}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial M}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial M}$$

$$\frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial \varepsilon}\Delta\varepsilon + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial F_s}\Delta F_s + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial M}\Delta M + \frac{\partial\left(\frac{\partial pdf}{\partial \alpha}\right)}{\partial \alpha}\Delta\alpha = -\frac{\partial pdf}{\partial \alpha}$$

In order to simplify the analytical evaluation of these equations good approximations can be determined from the pdf product equation by observing the effects of small changes in the individual parameters. As an example involving a small change in the detection efficiency, ε, the 1st order derivative

$$\left(\frac{\partial pdf}{\partial \varepsilon}\right) \approx \left(\frac{\delta pdf}{\delta \varepsilon}\right) = \frac{pdf\left(\varepsilon + \delta\varepsilon, F_s, M, \alpha, \mu_{R_1}, \mu_{R_2}, \mu_{R_3}\right) - pdf\left(\varepsilon, F_s, M, \alpha, \mu_{R_1}, \mu_{R_2}, \mu_{R_3}\right)}{\delta\varepsilon}$$

is arrived at. This expands to give the second order derivative approximation

$$\frac{\partial\left(\frac{\partial pdf}{\partial \varepsilon}\right)}{\partial F_s} \approx \frac{\delta\left(\frac{\delta pdf}{\delta \varepsilon}\right)}{\delta F_s} = \frac{\left(\frac{\delta pdf\left(\varepsilon, F_s, M, \alpha\right)}{\delta \varepsilon} - \frac{\delta pdf\left(\varepsilon, F_s + \Delta F_s, M, \alpha\right)}{\delta \varepsilon}\right)}{\delta F_s}$$

with the result that the linearisation equations are
approximated by

$$\frac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta\varepsilon}$$

$$\frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta F_s}$$

$$\frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta M}$$

$$\frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\varepsilon}\Delta\varepsilon + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta F_s}\Delta F_s + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta M}\Delta M + \frac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\alpha}\Delta\alpha = -\frac{\delta pdf}{\delta\alpha}$$

These equations are most readily handled in matrix notation;
D2 x Δ X = D1

$$\text{where } D1 = \begin{pmatrix} -\dfrac{\delta pdf}{\delta\varepsilon} \\ -\dfrac{\delta pdf}{\delta F_s} \\ -\dfrac{\delta pdf}{\delta M} \\ -\dfrac{\delta pdf}{\delta\alpha} \end{pmatrix}$$

$$D2 = \begin{pmatrix} \dfrac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta\varepsilon} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta F_s} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta M} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\varepsilon}\right)}{\delta\alpha} \\ \dfrac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta\varepsilon} & \dfrac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta F_s} & \dfrac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta M} & \dfrac{\delta\left(\frac{\delta pdf}{\delta F_s}\right)}{\delta\alpha} \\ \dfrac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\varepsilon} & \dfrac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta F_s} & \dfrac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta M} & \dfrac{\delta\left(\frac{\delta pdf}{\delta M}\right)}{\delta\alpha} \\ \dfrac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\varepsilon} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta F_s} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta M} & \dfrac{\delta\left(\frac{\delta pdf}{\delta\alpha}\right)}{\delta\alpha} \end{pmatrix}$$

$$\Delta X = \begin{pmatrix} \Delta\varepsilon \\ \Delta F_s \\ \Delta M \\ \Delta\alpha \end{pmatrix}$$

Successive estimates are given by $X_{n-1} = X_n + \Delta X_n$ which process
is repeated until the correction vector is below a certain
predetermined threshold which is accepted as negligible. A
final solution

$$\begin{pmatrix} \hat{\varepsilon} \\ \hat{F}_s \\ \hat{M} \\ \hat{\alpha} \end{pmatrix} = X_{final}$$

results. This solution provides a very good fit to the
experimental results obtained. From the spontaneous fission
rate value so obtained the mass of the neutron emitting
isotopes in the sample can be obtained. This in turn can be
linked to the overall neutron source mass.

In certain cases due to unusual sample conditions or due to
poor selection of the starting criteria the correction vector
provided may be such that a true solution cannot be obtained.
As illustrated in Figure 4a with the pdf 50 shown, the
differentiated pdf, Figure 5b, may not change sign between
the initial estimate 52 and the solution. If the initial
estimate 54 is fortunately located then a true solution 56
will be tended towards, but if the initial estimate 52 is not
so fortunate a false solution 58 will be tended towards.

To counter this a trial pdf is evaluated for both positive
and negative correction factors.

The correction vector may substantially overestimate or
underestimate the distance to a solution, because the
function is non-linear. To counter this the correction
vector for a pdf value is multiplied by 32 and then halved
and the pdf value recalculated. This pdf value is compared
against the original. If the new pdf value is less than the

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previous the process is repeated and the new value compared
with the new previous value. If the new value is greater
than the previous then the correction vector is applied at
that value. Otherwise the process is repeated until such a
situation is reached.

To avoid the situation where the initial pdf is zero and as a
consequence derivatives cannot be calculated, due to poorly
selected starting conditions for instance, the count rate
standard deviation is inflated in such a case.
Multiplication by a large constant is employed to this end.
After the process has converged towards a solution the value
of this factor is reduced in stages and the process repeated
until a solution is obtained with the inflationary factor set
at zero.

In any experimentally derived result it is important to know
the error possible in the result. This is particularly so
for the monitoring situations with which this method is
principally concerned as the implementation of the waste
disposal based upon it must always act on the worst possible
case in meeting the critical safety factors.

The associated error estimates are determined as follows. In
each case the precision of the final solution is dependant on
the precision of the count rates for the singlets, doublets
and triplets. Assuming these count rates are independent the
variance is given by

$$\left(\sigma_{\hat{\varepsilon}}\right)^2 = \left(\left(\frac{\partial\hat{\varepsilon}}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{\varepsilon}}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{\varepsilon}}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{F}_s}\right)^2 = \left(\left(\frac{\partial\hat{F}_s}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{F}_s}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{F}_s}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{M}}\right)^2 = \left(\left(\frac{\partial\hat{M}}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{M}}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{M}}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{\alpha}}\right)^2 = \left(\left(\frac{\partial\hat{\alpha}}{\partial R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\partial\hat{\alpha}}{\partial R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\partial\hat{\alpha}}{\partial R_3}\right)\sigma_{R_3}\right)^2$$

The partial derivatives again need not be determined but can be approximated by

$$\left(\frac{\partial \hat{\varepsilon}}{\partial R_1}\right) \approx \left(\frac{\delta \hat{\varepsilon}}{\delta R_1}\right) = \frac{\hat{\varepsilon}\left(\mu_{R_1} + \delta\mu_{R_1}, \mu_{R_2}, \mu_{R_3}\right) - \hat{\varepsilon}\left(\mu_{R_1}, \mu_{R_2}, \mu_{R_3}\right)}{\delta\mu_{R_1}}$$

where $\delta\mu_{R1}$ = a small change in μ_{R1} etc.
$\varepsilon(\mu_{R1}, \mu_{R2}, \mu_{R3})$ = final estimate from solution to $(\mu_{R1}, \mu_{R2}, \mu_{R3})$ count rates set with the partial derivatives being determined for each of the count rates based on the observed rates and small deviations in each.

The approximate error estimates are thus given by

$$\left(\sigma_{\hat{\varepsilon}}\right)^2 = \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{\varepsilon}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{F}_s}\right)^2 = \left(\left(\frac{\delta\hat{F}_s}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{F}_s}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{F}_s}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{M}}\right)^2 = \left(\left(\frac{\delta\hat{M}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{M}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{M}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

$$\left(\sigma_{\hat{\alpha}}\right)^2 = \left(\left(\frac{\delta\hat{\alpha}}{\delta R_1}\right)\sigma_{R_1}\right)^2 + \left(\left(\frac{\delta\hat{\alpha}}{\delta R_2}\right)\sigma_{R_2}\right)^2 + \left(\left(\frac{\delta\hat{\alpha}}{\delta R_3}\right)\sigma_{R_3}\right)^2$$

which in matrix notation becomes

$$VX = DR \times VR$$

$$\text{where } VX = \begin{pmatrix} \left(\sigma_{\hat{\varepsilon}}\right)^2 \\ \left(\sigma_{\hat{F}_s}\right)^2 \\ \left(\sigma_{\hat{M}}\right)^2 \\ \left(\sigma_{\hat{\alpha}}\right)^2 \end{pmatrix}$$

$$VR = \begin{pmatrix} \left(\sigma_{R_1}\right)^2 \\ \left(\sigma_{R_2}\right)^2 \\ \left(\sigma_{R_3}\right)^2 \end{pmatrix}$$

$$DR = \begin{pmatrix} \left(\dfrac{\delta\hat{\varepsilon}}{\delta R_1}\right)^2 & \left(\dfrac{\delta\hat{\varepsilon}}{\delta R_2}\right)^2 & \left(\dfrac{\delta\hat{\varepsilon}}{\delta R_3}\right)^2 \\[2mm] \left(\dfrac{\delta\hat{F}_s}{\delta R_1}\right)^2 & \left(\dfrac{\delta\hat{F}_s}{\delta R_2}\right)^2 & \left(\dfrac{\delta\hat{F}_s}{\delta R_3}\right)^2 \\[2mm] \left(\dfrac{\delta\hat{M}}{\delta R_1}\right)^2 & \left(\dfrac{\delta\hat{M}}{\delta R_2}\right)^2 & \left(\dfrac{\delta\hat{M}}{\delta R_3}\right)^2 \\[2mm] \left(\dfrac{\delta\hat{\alpha}}{\delta R_1}\right)^2 & \left(\dfrac{\delta\hat{\alpha}}{\delta R_2}\right)^2 & \left(\dfrac{\delta\hat{\alpha}}{\delta R_3}\right)^2 \end{pmatrix}$$

A series of experimental measurements were conducted to test
the performance of the maximum-likelihood multiplicity
analysis described above. These trials were carried out
using a passive neutron counting test rig containing 48 ^{3}He
neutron detector tubes encased in polythene moderator. The
chamber was sufficient to accommodate 200 litre drums and had
a neutron detecting efficiency of 15% for an empty drum and a
neutron die-away-time of 600m/s. A variety of matrix filled
drums were used in conjunction with Pu and ^{252}Cf standard
sources to simulate waste measurements.

In the first set of experimental trials, the reduction in
systematic errors (largely due to positional and matrix
affects) achieved by multiplicity counting compared with NCC
using a ^{252}Cf standard source to simulate a large quantity of
plutonium at various positions within waste matrix filled
drums was employed. The specified probability distributions
for this analysis were (ε, ε_0, ε_- = (4%, 10%, 16%), (M, M$_0$,
M$_+$) = 1.00, 1.00, 1.20) and (α, α_0, α_+) = 0.00, 0.00, 1.00).
These distributions were designed to represent a measurement
scenario where possibly large amounts of plutonium were
present within a matrix with highly variable neutron
moderating and/or absorption properties.

The results of these measurements (3000s count times) are
presented in Table 1. The first column of Table 1 gives
details of the matrix and the source position for each
measurement. The second column shows the measured net (i.e.

background corrected) neutron singles, doubles and triples
count rates and their associated standard deviations. The
third column gives the results of a maximum-likelihood
multiplicity analysis of the measured data. The fourth
column shows the results that have been obtained from a
conventional NCC analysis in which the measured doubles rate
is converted directly to a $^{240}Pu_{eff}$ mass by means of a
calibration factor. In this case, the calibration factor
assumes $\epsilon = 10\%$ (i.e. the most likely value of the
probability distribution specified for the multiplicity
analysis).

TABLE 1

Measurement Number	Net Count Rates /e -1	Multiplicity Results	NCC Results 240Pu$_{eff}$ / g
#1 PVC (85kg) source @ B	R1=22773 +/-3 R2=3471 +/-8 R3=269 +/-12	240Pu $_{eff}$=132.7g ϵ= 9.6% M= 1.00 α= 0.00	100.8g
#2 PVC (85kg) source @A	R1=20311 +/-3 R2=2778 +/-8 R3=185 +/-13	240Pu $_{eff}$=132.0g ϵ= 8.6% M= 1.00 α= 0.00	80.6g
#3 Steel (270kg) source @ B	R1=31861 +/-4 R2=6806 +/-12 R3=686 +/-26	240Pu $_{eff}$=132.2g ϵ= 13.5% M= 1.00 α= 0.00	197.6g
#4 Steel (270kg) source @ A	R1=37947 +/-4 R2=9674 +/-19 R3=1315 +/-39	240Pu $_{eff}$=132.1g ϵ= 16.0% M= 1.00 α= 0.00	280.8g

#5 PVC (50kg) source @ B	R1=28169 +/-3 R2=5344 +/-12 R3=484 +/-24	240Pu eff=131.9g e= 11.9% M= 1.00 α= 0.00	155.1g
#6 PVC (50kg) source @ A	R1=33051 +/-5 R2=7348 +/-13 R3=831 +/-28	240Pu eff=131.9g e= 14.0% M= 1.00 α= 0.00	213.3g
#7 Paper (20kg) source @ B	R1=28105 +/-5 R2=5304 +/-11 R3=511 +/-24	240Pu eff=132.3g e= 11.9% M= 1.00 α= 0.00	153.8g
#8 Paper (20kg) source @ A	R1=35463 +/-39 R2=8410 +/-22 R3=1091 +/-52	240Pu eff=132.9g e= 14.9% M= 1.00 α= 0.00	244.1g
#9 Water (105kg) source @ B	R1=20604 +/-10 R2=2856 +/-9 R3=181 +/-13	240Pu eff=132.1g e= 8.7% M= 1.00 α= 0.00	82.9g
#10 Water (105kg) source @ A	R1=13345 +/-2 R2=1200 +/-5 R3=51 +/-6	240Pu eff=131.8g e= 5.7% M= 1.00 α= 0.00	34.8g
#11 Polythene (22kg) source @ B	R1=29895 +/-4 R2=6015 +/-11 R3=569 +/-23	240Pu eff=131.7g e= 12.7% M= 1.00 α= 0.00	174.6g
#12 Polythene (22kg) source @ A	R1=34554 +/-21 R2=8004 +/-15 R3=992 +/-34	240Pu eff=132.5g e= 14.6% M= 1.00 α= 0.0	232.4g

The $^{240}Pu_{eff}$ masses determined by the two techniques are
compared graphically in Figure 5. The arrow bars shown on
the multiplicity results have be determined by the maximum-
likelihood analysis and include both the systematic and
random components. The arrow bars shown on the MC results
also include both random and systematic components. The
systematic term has been determined by assuming the same
probability distribution for the detection efficiencies were
specified for the multiplicity analysis and the random term
is simply the standard deviation on the measured doubles
rate.

The consistently accurate results obtained from the maximum-
likelihood MC analysis have been obtained by (correctly)
varying ϵ (rather than M and α) to match the predicted and
measured count rates. This is true even in extreme cases
such as the measurement number 10 where the source was placed
at the centre of a water-filled drum.

In a second set of tests a small Pu source (nominal $^{240}Pu_{eff}$
mass = 0.42g) was measured under similar conditions to
establish what benefits the maximum-likelihood MC analysis
would provide for the measurement of smaller quantities of
Pu. The specified probability distributions for the maximum
likelihood MC analysis were (ϵ, ϵ_0, ϵ_+ = (10%, 13%, 16%), (M,
M_0, M_+) = 1.00, 1.00, 1.01) and (α, α_0, α_+) = 1.40, 1.70,
2.00). The relatively tight distribution specified for the M
reflects the fact that for small quantities of Pu, it is safe
to assume that M will be close to unity. The relatively wide
distribution specified for α reflects the fact that this
parameter will be difficult to estimate for real wastage of
variations in chemical composition and impurity content.

The results of these measurements (3000s count times) are
presented in Table 2 and a comparison of the $^{240}Pu_{eff}$ mass is

determined by the maximum-likelihood MC and NCC techniques
are illustrated in Figure 6.

TABLE 2

Measurement Number	Net Count Rates /ϵ -1		Multiplicity Results	NCC Results 240Pu$_{eff}$ / g
#1 PVC (85kg) source @ A	R1=124.34 R2=3.88 R3=0.32	+/-0.31 +/-0.08 +/-0.05	240Pu $_{eff}$=0.40g ϵ= 10.3% M= 1.00 α= 1.93	0.26g
#2 Wood (48kg) source @ B	R1=125.93 R2=4.50 R3=0.29	+/-0.34 +/-0.07 +/-0.03	240Pu $_{eff}$=0.36g ϵ= 11.8% M= 1.00 α= 1.89	0.30g
#3 Wood (48kg) source @ A	R1=156.94 R2=7.10 R3=0.69	+/-0.33 +/-0.10 +/-0.10	240Pu $_{eff}$=0.41g ϵ= 13.8% M= 1.00 α= 1.70	0.47g
#4 PVC (50kg) source @ B	R1=121.46 R2=4.41 R3=0.48	+/-0.33 +/-0.08 +/-0.14	240Pu $_{eff}$=0.38g ϵ= 11.3% M= 1.00 α= 1.76	0.29g
#5 PVC (50kg) source @ A	R1=159.82 R2=7.09 R3=0.61	+/-0.34 +/-0.09 +/-0.04	240Pu $_{eff}$=0.38g ϵ= 14.2% M= 1.01 α= 1.92	0.47g
#6 Paper (20kg) source @ B	R1=137.75 R2=5.62 R3=0.47	+/-0.34 +/-0.07 +/-0.05	240Pu $_{eff}$=0.37g ϵ= 13.0% M= 1.00 α= 1.79	0.37g
#7 Paper (20kg) source @ A	R1=173.61 R2=8.77 R3=0.86	+/-0.35 +/-0.10 +/-0.06	240Pu $_{eff}$=0.39g ϵ= 15.5% M= 1.00 α= 1.83	0.58g

#8 Polythene (22kg) source @ B	R1=133.44 R2=5.22 R3=0.66	+/-0.33 +/-0.09 +/-0.16	240Pu $_{eff}$=0.40g ϵ= 12.0% M= 1.00 α= 1.72	0.35g
#9 Polythene (22kg) source @ A	R1=172.47 R2=8.09 R3=0.84	+/-0.36 +/-0.11 +/-0.09	240Pu $_{eff}$=0.43g ϵ= 14.3% M= 1.00 α= 1.73	0.54g

As can be seen from Figure 6, the maximum-likelihood MC
analysis results are again consistently closer to the correct
value than the conventional NCC results. This emphasises the
ability of the maximum-likelihood technique to find the
correct solution to the multiplicity equations without
requiring any of the unknown parameters to be specified
exactly.

The fact that the arrow bars shown for the MC analysis of the
Pu source is significantly larger than those for the ^{252}Cf
measurements reflects the fact that the relative standard
deviations on the measured count rates for the far smaller Pu
source are significantly greater than those of the ^{252}Cf
source.

A third set of measurements were also made on drums filled
with inactive matrices to evaluate the improvement in the
limit of detection that would result from the use of the
maximum-likelihood analysis technique compared with NCC.
Several sets of hour long measurements were performed on the
matrix filled drums and similar measurements were made with
an empty chamber to determine the ambient background. The
net count rates were determined and then analysed by both
maximum-likelihood MC and conventional NCC techniques. The
specified probability distributions for the maximum-
likelihood MC analysis were (ϵ, ϵ_0, ϵ_+ = (10%, 13%, 16%), (M,
M_0, M_+) = 1.00, 1.00, 1.01) and (α, α_0, α_+) = 1.40, 1.70,
2.00).

The comparisons of the results obtained from the two
techniques are shown graphically in Figure 7 for a drum
containing 50kg of scrap PVC and Figure 8 for a drum
containing 270kg of steel hulls. Also shown on the graphs is

an indication of the TRU/LLW segregation boundary at 100nCi/g
(calculated by assuming $94\%^{235}Pu$; $6\%^{240}Pu$).

The NCC results in Figure 7 are scattered either side of zero
because they depend solely on the variation in the measured
doubles rate from the matrix drum to the empty chain of
measurements. The multiplicity results, as discussed
earlier, have been constrained to the physically meaningful
situation of $^{240}Pu_{eff}$ greater or equal to zero. More
specifically, the magnitude and the measurement uncertainty
on the multiplicity results is consistently less than half
that of the NCC results. This translates directly to a
limited protection for the maximum-likelihood MC technique
which is less than half that of a conventional NCC analysis
performed under the same measurement conditions. This
improvement over NCC detection limits is as expected given
that more useful information is incorporated into the
maximum-likelihood MC analysis.

The results as shown in Figure 8 for the drum containing the
steel again show reduced measurements uncertainties for the
MC results compared with the NCC results, but now show
significant positive bias in the assay results as evident
despite the evidence of any spontaneous fissile material.
Cosmic ray induced neutrons (which are generated primarily in
high Z materials) are the likely cause of this affect. At
its current level the bias still allows re-categorisation
against 100nCi/g LLW limit. This is despite the lack of
background shielding on experimental systems without the use
of matrix specific background measurements.

The improvement in limit of detection shown in Figures 6 and
7 indicates that the maximum likelihood multiplicity analysis
technique will provide a consistent and reliable method for
the segregation of TRU/LLW at the 100nCi/g boundary.

The present invention therefore provides a technique whereby the unknown parameters for a waste sample can be determined with far greater accuracy whilst avoiding undue assumptions about the system. In waste investigations accurate Pu mass measurements can be made over the full range of wastes down to and below the 100nCi/g LLW boundary. The result is a more accurate and cost effective monitoring procedure with advantages in inventory control and with significant cost savings in organising waste disposal. The technique allows the low cost and high reliability associated with passive neutron counting systems to be retained. These advantages are obtained despite variations in the make-up of the waste and in the location of the neutron source within that waste. This improved monitoring is backed up by a firm indication of the errors encountered. The parameter envelopes applied to the various functions can be updated as the monitoring of samples progresses. Thus the results from previous samples can improve the modelling of subsequent samples in the waste stream.